

July 2, 1999

MEMORANDUM

SUBJECT: Dimethoate (PC Code 35001; CAS # 60-51-5): Interim Memorandum
on the Effect of Peeling, Washing or Cooking on Concentrations of
Dimethoate in Foods.

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Summary

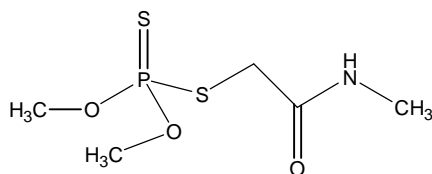
A search of the literature (1967 to present) was undertaken to identify information pertaining to the effects of kitchen processing (e.g. washing, peeling, cooking) on the concentration of dimethoate residues in foods. Such information could be potentially useful for assessing dietary exposure to dimethoate. A total of eight studies were identified. The studies investigated the effect of kitchen-type processing on dimethoate residues in several different types of vegetables that included: potatoes, cabbage, green peas, carrots, cauliflower, *Brassica rapa*, and rice. (No studies were identified that pertain to the effect of kitchen processing on residues of dimethoate in fruits, milk, beef, pork, poultry, eggs, or other non-vegetable commodities.) The results of these studies are described herein. To summarize the findings of these studies, all of the studies show that kitchen processing causes a decrease in the concentrations of dimethoate residues in vegetables. The decrease ranged from about 20% to 91%, depending upon the vegetable and the type and extent of processing used. The results from these studies should be useful in the estimation of dietary exposure to dimethoate. Many of these studies investigated other organophosphorus pesticides as well, and some of the results pertaining to the other organophosphorus substances are also discussed.

Background

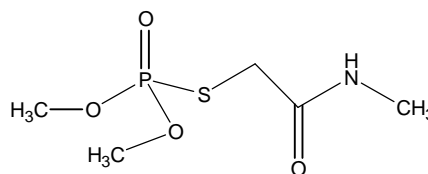
Dimethoate is a thio-organophosphorus insecticide registered for use in the United States to prevent or treat insect infestations on a wide variety of crops. These crops include, for example, oranges, broccoli, apples, cauliflower, wheat, rice and cotton, to name just a few. In neat form, dimethoate is a crystalline, low melting solid (mp = 52-52.5 °C), having a water solubility of about 6 grams/liter at 25 °C. Dimethoate has approximately 6-fold greater affinity for lipids than it has for water. [Its octanol-water partition coefficient (log P) is about 0.78.] Like many other organophosphorus insecticides, dimethoate is an irreversible inhibitor of acetyl cholinesterase. This property serves as the basis of its insecticidal action, and is believed to be at least partly responsible for causing neurotoxic effects in mammals.

In animals and plants, dimethoate is metabolized to an oxon metabolite. In this metabolic conversion the doubly-bonded sulfur atom (=S) of dimethoate is replaced with an oxygen atom. The oxon metabolite is also capable of irreversibly inhibiting acetyl cholinesterase. The structures of these substances are shown the following page. Tolerances are established for combined residues of dimethoate and its oxon metabolite, and range from 0.002 ppm to 2 ppm.

As part of its process to assess the risks posed by pesticides, EPA's Office of Pesticide Programs (OPP) estimates dietary exposure to pesticides that are registered for use in or on agricultural commodities or that otherwise may be present in foods. Dietary exposure to a pesticide includes exposure to the parent substance and any metabolites or degradates of toxicological concern. Data needed for estimation of the levels of pesticide residues in foods as consumed are derived or obtained from several sources that include supervised field trials, residue monitoring programs, and metabolism and food processing studies. Generally, the concentration



Dimethoate



Oxon metabolite of dimethoate

of pesticide residues in a food at the time of consumption are much lower than the concentration of residues in the raw agricultural commodity from which the food was obtained. This is because many raw agricultural commodities undergo some type of processing after harvest and prior to consumption. Examples of types of processing that may take place in the home or elsewhere, and cause declines in residue concentration include: trimming; washing; peeling; and cooking. While data from field trials are very useful in estimating dietary exposure, these types of data typically do not represent the influence of typical kitchen-type processing (i.e., trimming, washing, peeling, or cooking) on the levels of pesticide residues in foods. Studies that are designed specifically to measure the effect of trimming, washing, peeling, or cooking on the concentration of pesticide residues in a given food are particularly useful in combination with data from field trials and residue monitoring programs when more accurate or refined dietary exposure estimates are warranted.

Some of the crops on which dimethoate is registered for use are typically washed, trimmed, peeled or cooked prior to consumption. Hence, consideration of the effect of these types of kitchen-type processing on the levels of dimethoate residues in foods should be undertaken in order to accurately estimate dietary exposure to dimethoate. However, no data pertaining to the effect of on the concentrations of dimethoate residues in foods have been submitted to OPP. In an attempt to fill this data gap, a search of the literature (from 1967 to present) was conducted by OPP to identify such data. Eight studies were identified. While all of the literature articles pertaining to the studies are discussed herein, only three of the articles could be obtained at the time of this writing. Due to the urgency of this matter, this memo was prepared without full review of some of the studies identified. Data mentioned herein pertaining to studies not yet received came from the abstracts of the studies, and are indicated as such. This memorandum is intended to serve only as an **interim** memorandum until the remaining literature studies have been received and reviewed, at which time this memorandum will be modified accordingly.

Results

A total of eight studies describing the effect of kitchen processing (i.e., washing, peeling or cooking) on dimethoate residues in foods were identified (1-8). Each of these studies pertain to vegetable commodities: no studies were identified that pertain to residues of dimethoate in fruits, milk, beef, pork, poultry, eggs, or other non-vegetable commodities. The results of these

studies are summarized below. Three of the literature articles (1-3) pertaining to the studies were obtained. Information from the remaining five studies (4-8) were obtained from the abstracts of the literature articles. Most of the eight studies describe the effect of kitchen processing on concentrations of residues of other organophosphorus pesticides as well.

Several of the studies investigated the effect of kitchen-type processing on dimethoate and other organophosphorus pesticides residues in potatoes, cabbage, green peas or carrots. In a study conducted by Askew, et al. (1), potatoes or cabbage (50 g) were macerated with 200 ml of water for 5 minutes, fortified with 2 ppm pesticide, and diluted to 300 ml with water. The mash was boiled gently for 30 minutes, then cooled rapidly and centrifuged. A 150 ml aliquot of supernatant liquid was extracted with 100 ml chloroform and 40 ml of the chloroform layer was dried and evaporated to dryness. The remaining residue was dissolved in acetone and the pesticide content determined by gas chromatography. For dimethoate, the percentage hydrolysis in potato mash ranged from 32-39%, and in cabbage mash ranged from 44-52%. Hydrolysis data in potato mash and cabbage mash for the other organophosphorus pesticides tested are as follows: chlorfenvinphos, 37-53%, 56-80%; methyl parathion, 59-62%, 76-78%; ethyl parathion, 49-64%, 91-93%; and phorate 100%, 100%, respectively.

Sugibayashi, et al (4) studied and compared the effects of washing, peeling, and cooking on the residual levels of dichlorvos, dimethoate and other pesticides in white potatoes and carrots. In this study the outer surfaces of the vegetables were experimentally treated with pesticide. The level of pesticides did not significantly differ between that of vegetables after simple water washing and that after detergent washing. Peeling was the most effective way to remove the pesticide from the vegetables, and the second most effective way was frying. Boiling was effective in reducing the level of the more water soluble pesticides, and the use of oil in cooking processes markedly decreased the level of the more fat soluble pesticides. No specific or quantitative details on dimethoate are provided in the study abstract.

In a study conducted by Wen, et al. (5), chinese cabbage, potatoes, green peas, and carrots were chopped and immersed for 30 minutes in water containing 1000 microgram/mL of dimethoate. The vegetables were either washed, boiled, fried, baked, or soaked with vinegar or alcohol and dimethoate residues were measured. This experiment was repeated using chlorfenvinphos-E, S-benzyl diisopropyl phosphorothiolate, monocrotophos-E, diazinon, dieldrin, fenitrothion, dichlorvos, and chlorothalonil. Chlorfenvinphos-E, S-benzyl diisopropyl phosphorothiolate, and monocrotophos-E were “efficiently” removed by washing alone. In Chinese cabbage boiled for 30 minutes, decomposition of diazinon, dieldrin, dimethoate, fenitrothion, and chlorothalonil ranged from 72-99%. (It is not discernable from the study abstract where the decomposition of dimethoate falls within this range.) Only diazinon did not dissolve in soaking vinegar. Emulsifiers contained in the pesticides did not affect the removal of the pesticides. There were correlations between chlorfenvinphos-E, diazinon, or dichlorvos in Chinese cabbage and washing, washing with detergent, and boiling treatments, and between dimethoate and soaking with 40% ethyl alcohol. No additional details are provided in the study abstract.

One of the studies (7) investigated the effect of kitchen processing on residues of diazinon and dimethoate in green beans and cauliflower grown under usual growing conditions and treated with typical applications of these pesticides. In the vegetable samples that had been pesticide-treated under usual growing conditions, the residues of diazinon and dimethoate ranged from 0.011-0.026 ppm and 0.012-0.056 ppm, respectively. An insecticide treatment shortly before harvest (3 or 7 days earlier) caused a marked increase, depending on the type of vegetable, in residues (0.076-0.130 ppm of diazinon and 0.060-0.290 ppm of dimethoate). Kitchen-processing techniques (cleaning, peeling, washing, cooking removed 40-77% of the diazinon residues, and 37-82% of the dimethoate residue in the vegetables examined. No additional details are provided in the study abstract.

Khaire and Dethe (6) investigated the presence of dimethoate residues in unprocessed and processed cauliflower curds previously treated with 0.03 and 0.06% dimethoate spray. Analyses of residues was accomplished by microbioassay and chemoassay. Washing alone reduced the levels of dimethoate residues by approximately 25-80%, whereas washing and cooking of curds reduced the level of residues by 52-91%. No specific details regarding duration or temperature of cooking are provided in the study abstract.

Ishikura and co-workers (2) studied the effect of boiling on residues of pesticides in rice. The pesticides tested include dimethoate, phosdrin, EPN (phenylphosphonothioic acid O-ethyl O-(4-nitrophenyl) ester), methyl parathion, phorate, ethion, and ronnel. The experimental procedure involved washing 200 grams of polished rice in distilled water. The water is drained, and 300 ml of distilled water and 0.4 ml of a methanol solution containing a single pesticide at a concentration of 1000 ppm were added to the rice. The rice was boiled in this mixture using an electric rice cooker. The total cooking time was 18 minutes (heating time after the water started to boil was about 15 minutes). Residual pesticide concentrations in the rice ranged from 20-94% after boiling. The specific concentrations of residues of each pesticide remaining in the rice are as follows: dimethoate, (80.0%); phosdrin (61.8%); EPN (53.8%); methyl parathion (42.1%); ethion (9.6%); phorate (8.9%); and ronnel (5.5%). Hence, 20% of the initial amount of dimethoate was decomposed by boiling.

In a study conducted by Schattenberg, et al. (3) 17 types of raw fruits and vegetables frequently consumed by children and grown in the United States were obtained in 10 lb quantities by random sampling from different shipment areas. A 5 lb portion of each sample were analyzed for pesticide residues in accordance to U.S. FDA procedures (Pesticide Analytical Manual, Vol. I). In samples where residues were found, the remaining 5 lbs were analyzed after being subjected to kitchen-type processing (washing, peeling, and/or cooking). Low levels of pesticide residues were detected in 97 (40%) of 243 samples analyzed. The number of samples containing detectable residues dropped to 47 (19%) after household preparation. Results indicate that residue levels in most commodities are substantially reduced after typical household preparation. Of the 243 samples analyzed, only green beans contained detectable residues of dimethoate. These residues were found in 0.19 ppm. (The tolerance of dimethoate on green beans is 2 ppm.) Processing of the green beans consisted of removal of stems, washing with dilute surfactant, rinsing in water, and boiling for 5 minutes. Residues after processing were below 0.15 ppm.

Watanabe, et al. (8) investigated the stability of dimethoate in vegetables of the *Brassica rapa* type. (The specific vegetables used in this study are not mentioned in the abstract, but examples of vegetables belonging to this class include turnips, mustard spinach, mizuna). The vegetable samples contained 0.020-0.070 ppm of dimethoate. (It is not stated in the abstract how the dimethoate was applied onto the vegetables.) The vegetables were boiled in water for 10 minutes. Residues of dimethoate were reduced by approximately 50%.

Discussion

The studies described above investigated the effect of kitchen-type processing on the concentration of dimethoate residues in several different types of vegetables that included: potatoes, cabbage, green peas, carrots, cauliflower, *Brassica rapa*, and rice. The studies all indicate that kitchen processing of vegetables that contain residues of dimethoate will cause decreases in the concentrations of the residues. While the results from these studies were summarized mostly from the study abstracts, and specific details of the results are not always provided in the abstracts, there appears to be general consistency among the overall results from all studies regarding the behavior of dimethoate residues during processing.

The study results are also in generally good agreement with what one would intuitively expect to happen to the concentration of dimethoate residues in a vegetable following kitchen processing. For example, peeling or washing a vegetable to which dimethoate was previously applied onto its outer surfaces is expected to result in a large decline in residues. Removal of the outer surfaces would obviously remove essentially all of the residues. Thorough rinsing of the vegetable under tap water is expected to greatly reduce the concentration of residues since dimethoate has appreciable solubility in water. Peeling or washing the outer surfaces would not remove residues that are inside (below the outer surface) of the vegetable. Many of the studies show that dimethoate residues in vegetables decrease upon cooking. Dimethoate is a low melting, low molecular weight phosphate ester that contains several different heteroatoms and that can react with various nucleophiles. It is not surprising, therefore, that it is not stable during kitchen processing that involves heat.

While all the studies show a decline in the concentration of dimethoate residues following processing of contaminated vegetables, there appear to be some differences between the study results regarding the extent of residue decline in the same vegetable. The study by Askew, et al. (1), for example, showed that 30 minutes of boiling cabbage fortified with 2 ppm of dimethoate caused a 44-52% reduction in dimethoate residues. In the Wen study (5), boiling of dimethoate-contaminated cabbage for 30 minutes caused at least a 72% decrease in the concentration of dimethoate residues. These differences may be attributable to differences in study design. The results from all of the studies suggest that there may be differences in the stability of dimethoate residues in different vegetables. For example dimethoate residues appear to be more stable in rice than they are in vegetables of the *Brassica rapa* type. In the Ishikura study (2), boiling rice contaminated with dimethoate caused only a 20% reduction in the concentration of residues after about 15 minutes. (Boiling for a longer period would likely cause further

reduction). However only 10 minutes of boiling of dimethoate-contaminated vegetables of the *Brassica rapa*-type caused a 50% reduction of residues (8). These apparent differences in dimethoate stability in different vegetables could be due to the biological differences between the vegetables or differences in study design.

The results of these studies show that the extent of decline in the concentrations of dimethoate residues in vegetables is greatly dependent upon the type of kitchen processing employed. Sugibayashi, et al (4) studied and compared the effects of washing, peeling, cooking on the residual levels of several pesticides (including dimethoate) in white potatoes and carrots. In this study the outer surfaces of the vegetables were experimentally treated with pesticide. Of the three different types of processing used in the study, peeling was the most effective way to remove the pesticide from the vegetables, and the second most effective way was frying. Boiling was effective in reducing the level of the more water soluble pesticides, and the use of oil in cooking processes markedly decreased the level of the more fat soluble pesticides. Thus, in addition to heat itself, the source of the heat (boiling versus frying), as well as water and fat solubility are important considerations that govern removal of pesticide residues from a food during cooking. Frying is often conducted at temperatures that are higher than those needed for boiling, which may explain why it caused a greater decline in dimethoate residues than did boiling. It should be noted that specific or quantitative details on dimethoate are not provided in the study abstract. Dimethoate is soluble in both fats and lipids, and has approximately 6 times the affinity for fat than it has for water. Cooking processes that involve either boiling or use of cooking oils should cause declines in dimethoate residues. It appears that use of more than one type of processing may cause greater decline of dimethoate residues than use of either type of processing alone. Khaire and Dethe (6) observed that washing alone reduced the levels of dimethoate residues by approximately 25-80% in cauliflower curds sprayed with dimethoate, whereas both washing and cooking of curds reduced the level of residues by 52-91%. (No specific details regarding duration or temperature of cooking are provided in the abstract.)

Use of Data Described Herein for Assessing Dietary Exposure to Dimethoate

The results from the studies described in this memorandum can be useful for making qualitative and semi-quantitative judgements regarding dietary exposure to dimethoate. Below are some guidelines for making such judgements.

Vegetable Crops

For vegetable crops in which dimethoate has been applied to the outer surface, peeling of such crops can be expected to result in essentially complete (i.e., 90-100%) removal of dimethoate residues. Washing with water will also cause a large reduction in dimethoate residues. The efficiency of removal of dimethoate residues from the outer surfaces by washing is dependent upon how thoroughly the washing is conducted. In the studies described above, it appears that washing alone accounted for a 25 to 80% (cauliflower curds) loss of residues. Peeling or washing

will obviously not remove residues that are not confined to the outer surface of a vegetable. Such residues would include those that are within the matrix of the vegetable. Thus, for vegetables in which dimethoate has been applied externally, and that are known or likely to be peeled prior to consumption, one should assume 100% removal of dimethoate residues. If the vegetable is not likely to be peeled prior to consumption, one can assume anywhere from 25 to 80% removal of the dimethoate residues if the vegetable is at least washed with water prior to consumption.

Cooking of vegetables that contain residues of dimethoate (either internally or externally) will cause the concentrations of the residues to decline. It appears from the data described above that the extent of decomposition of residues is dependent upon several factors that include the type of cooking (i.e., boiling versus frying), the duration of cooking, and the type of vegetable. Dimethoate residues decline by about 50% in *Brassica rapa* type vegetables (e.g., turnips, mustard spinach, mizuna) following boiling in water for 10 minutes. Dimethoate residues decline only by about 20% in rice boiled for 15 minutes. In cabbage and potatoes, dimethoate residues decline by about 48% and 35%, respectively, following 30 minutes of boiling. These boiling times for these vegetables are similar to those used during home boiling of these vegetables, and it is recommended that the corresponding decline values be used for estimating dietary exposure.

Frying appears to cause greater decline in dimethoate residues than does boiling. This is likely to be due to the higher temperatures often involved with frying. No quantitative data on the effect of frying on dimethoate residues are available from the study abstracts. As surrogate data for frying, one should consider using residue decline data from cooking studies in which vegetables were boiled, and increasing these data by 20%. The 20% increase is intended to account for the influence of increased temperature during frying and the fact that frying is often conducted in oil.

For vegetables treated externally with dimethoate and that are likely to undergo more than one type of processing prior to consumption, such as washing followed by cooking, one could assume at least 75% removal of dimethoate residues.

Non-Vegetable Crops

None of the studies described above contain information regarding the effect of processing on dimethoate residues in non-vegetable crops. Nonetheless, the information pertaining to vegetable crops may in some instances be used as surrogate data for non-vegetable crops, as described below.

Fruits

It is logical to assume that residues of dimethoate on the surfaces of fruits would behave similarly during processing to those found on the surfaces of vegetables. Hence, for fruits crops in which dimethoate has been applied to the outer surface, peeling of such crops can be expected to result in essentially complete (i.e., 90-100%) removal of dimethoate residues. Washing with water will also cause a large reduction in dimethoate residues. The efficiency of removal of dimethoate residues from the outer surfaces by washing is dependent upon how thoroughly the

washing is conducted. Peeling or washing will obviously not remove residues that are not confined to the outer surface of a fruit. Such residues would include those that are within the matrix of the fruit. Thus for fruits in which dimethoate has been applied externally, and that are known or likely to be peeled prior to consumption, one should assume 100% removal of dimethoate residues. If the fruit is not likely to be peeled prior to consumption, one can assume anywhere from 25 to 80% removal of the dimethoate residues if the fruit is at least washed with water prior to consumption.

Meat (beef, pork, poultry)

No studies that investigated the effect of cooking on dimethoate residues in meats were identified. A study was identified, however, that investigated the stability of other thio-organophosphorus insecticides in beef during cooking (9). This study investigated the stability of five thio-organophosphorus insecticides that included: famphur; fenthion; coumaphos; ronnel; chlorpyrifos; and one organophosphorus insecticide (stirofos oxon) in ground beef cooked at an internal temperature of 77 °C (approximately equal to 171 °F) and maintained at pHs of either 4.5, 5.5 or 6.5. The cooking temperature of 77 °C was selected because this temperature falls within the normal cooking range of chopped meat. The meat samples were heated in a water bath maintained at 81 °C. The time necessary for the internal meat temperature to reach 77 °C was 7 minutes and 19 seconds. Higher internal temperatures were not tested. The pH's of 4.5 and 5.5 were selected because these pHs fall within the range commonly encountered in fermented beef summer sausages. The selection of pH 6.5 was based on a report that the pH of muscle foods ranges from 5.4 to 7.0 .

The results of this study are summarized briefly here. In beef samples fortified individually with the substances tested, it was shown that all parent substances are stable in unheated meat at pH 4.5 and a temperature of 21 °C, but the stability declined as pH became more neutral. Mean percent recoveries of coumaphos, for example, in unheated meat at 21 °C were as follows: 84.5% (pH 4.5); 78.3% (pH 5.5); 67.2% (pH 6.5). Heating caused additional reduction in the concentration of all parent substances. With coumaphos, mean percent recoveries in beef heated to an internal temperature of 77 °C (approximately equal to 171 °F) were as follows: 55.3% (pH 4.5); 64.6% (pH 5.5); 61.0% (pH 6.5). Primary and secondary metabolites of coumaphos were identified in the beef samples post-heating. These metabolites, believed to be the oxygen analog of coumaphos and 3-chloro-4-methyl-7-hydroxycoumarin, respectively, were found in very low concentrations. Consequently, accurate identification and quantification was not possible. Similar findings occurred with the other substances tested in the study. Of all of the substances tested, the least stable was ronnel. Mean percent recoveries for ronnel were as follows: 43.7% (pH 4.5); 40.4% (pH 5.5); 24.4% (pH 6.5). The most stable substance was coumaphos.

Essentially all meats (beef, pork, poultry) consumed in the United States are cooked prior to consumption. In recent years much public concern has been given to the presence of potentially harmful bacteria in meats. To ensure food safety, food processing facilities such as restaurants are now required to cook meats at temperatures much higher and for durations longer than those used in the past. It is also recommended that these temperatures and durations be used when meats are cooked at home. Results from the study described above indicate that thio-

organophosphorus substances are labile (i.e., decompose) in beef when cooked, especially as pH becomes more acidic. Although dimethoate was not tested, it is logical to assume that it too is labile in beef during cooking since it is a thio-organophosphorus substance and it is known to decompose in vegetables during cooking. While higher internal temperatures and longer heating intervals were not tested in the above beef study, it is reasonable to expect that higher internal temperatures and/or longer heating intervals such as those that often occur during others types of cooking of beef (e.g., oven roasting, boiling, charcoal grilling) will cause greater reductions in the concentrations residues in beef.

It is recommended that a reduction factor of at least 40% be used for estimating dietary exposure to dimethoate residues in cooked meats. This factor is based on the stability of coumaphos in the beef cooking study described above.

Milk

Essentially all milk consumed in the United States is pasteurized prior to consumption. Pasteurization is the process of heating milk to kill pathogenic bacteria, and most other bacteria, without greatly altering flavor. No studies were found regarding the effect of pasteurization on the stability of dimethoate residues in milk. In the United States, pasteurization is carried out by batch- or continuous-flow processes that involve heating milk. (The U.S. Public Health Service has described their regulatory guidelines for proper milk pasteurization in the U.S. Public Health Service Pasteurized Milk Ordinance document.) In the batch process, each particle of milk must be heated to at least 63 °C and held continuously at this temperature for at least 30 minutes. In the continuous process, milk is heated to at least 72 °C for at least 15 seconds (this is often referred to as high-temperature-short time (HTST) pasteurization). Another continuous pasteurization process, known as ultrahigh temperature pasteurization, involves a shorter time (2 seconds) and a higher temperature (minimum of 138 °C).

Although no studies were found regarding the effect of pasteurization on the stability of dimethoate residues (or other OPs) in milk, the results from the cooking studies investigating the stability of dimethoate and other OPs in vegetables can provide some insight into the stability of dimethoate in milk during pasteurization by the batch process. The vegetable cooking studies and the batch pasteurization studies involve heating for several minutes or longer. It appears quite possible that dimethoate residues are labile under the conditions of batch process pasteurization. Some caution needs to be exercised, however, in basing judgements regarding the stability of dimethoate residues in milk during pasteurization on the vegetable cooking studies described above. While the batch process involves heat, it requires a minimum temperature of 63 °C, which is considerably lower than the temperatures (about 100 °C) used in the studies in which vegetables were boiled. Both the HTST and the ultrahigh temperature pasteurization processes use considerably shorter heat durations compared to the batch process. Hence, while it seems reasonable to assume that dimethoate residues in milk will decompose during pasteurization, there are not enough data available to estimate quantitatively the stability of dimethoate during pasteurization. It is recommended that milk data from the USDA's Pesticides Data Program be used, if available, because these data would reflect the effect of pasteurization on dimethoate

residues.

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